Supporting Information

Molecular Self-Assembly under Nanoconfinement: Indigo Carmine Scroll Structures Entrapped Within Polymeric Capsules

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Figure S1. Cryo-TEM image of the polymeric nanoparticles depicting nanocapsules and nanospheres at low (A) and high (B) magnifications, corresponding histogram for size distribution (C).



Figure S2. UV-visible spectra. (A) of **IC** (1) and **IC** encapsulated within the polymeric nanocapsules (2) samples in water. Inset: Photos showing the corresponding two samples (1 and 2). (B) Zooming into the region (450-750 nm).

Specimen preparation

A small droplet of the solution was placed on a carbon-coated perforated polymer film supported on a 200-mesh TEM copper grid, held by tweezers. The drop was turned into a thin film (preferably less than 300 nm) by blotting away excess solution with a metal strip covered with a filter paper. The grid was then plunged quickly into liquid ethane at its freezing point (-183 °C) to ensure its vitrification (amorphous ice formation) and to better preserve the suspended nanostructures. The vitrified specimen was transferred under liquid nitrogen into a Gatan 626DH cryo-holder and equilibrated below -170 °C.

Captions for Supplementary Movies

Supplementary Movie 1: cryo-TEM tomography results following segmentation and visualization with Amira software. The movie depicts the 3D structure of the encapsulated **IC** in the polymeric nanocapsule as reconstructed from a tilt-series cryo-TEM tomography experiment. The missing parts at the top and bottom of the capsule result from the missing wedge of the tilt-series ((-60°) to 60°). (MP4).

Supplementary Movie 2: cryo-TEM tomography results following segmentation and visualization with Amira software at a perpendicular angle compared to movie 1. The movie depicts the 3D structure of the encapsulated IC in the polymeric nanocapsule as reconstructed from a tilt-series cryo-TEM tomography experiment. It presents well, the folding of the **IC** molecules inside the capsule. The missing parts at the top and bottom of the capsule result from the missing wedge of the tilt-series (-60° to 60°). (MP4).

Supplementary Movie 3: Tomographic tilt series of the encapsulated **IC** in the polymeric nanocapsules. (MP4).